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ABSTRACT

The accuracy associated with assessing the environmental consequences of an accidental release of radioactivity is highly dependent on our knowledge of the source term characteristics and, in the case when the radioactivity is condensed on particles, the particle size distribution, all of which are generally poorly known. This paper reports on the development of a numerical technique that integrates the radiological measurements with atmospheric dispersion modeling. This results in a more accurate particle size distribution and particle injection height estimation when compared with measurements of high explosive dispersal of ^{239}Pu .

The estimation model is based on a non-linear least squares regression scheme coupled with the ARAC three-dimensional atmospheric dispersion models. The viability of the approach is evaluated by estimation of ADPIC model input parameters such as the ADPIC particle size mean aerodynamic diameter, the geometric standard deviation, and largest size. Additionally we estimate an optimal "coupling coefficient" between the particles and an explosive cloud rise model. The experimental data are taken from the Clean Slate 1 field experiment conducted during 1963 at the Tonopah Test Range in Nevada.

The regression technique optimizes the agreement between the measured and model predicted concentrations of ^{239}Pu by varying the model input parameters within their respective ranges of uncertainties. The technique generally estimated the measured concentrations within a factor of 1.5, with the worst estimate being within a factor of 5, very good in view of the complexity of the concentration measurements, the uncertainties associated with the meteorological data, and the limitations of the models. The best fit also suggest a smaller mean diameter and a smaller geometric standard deviation on the particle size as well as a slightly weaker particle to cloud coupling than previously reported

1. INTRODUCTION

During the reduction of world-wide nuclear weapons stockpiles, the required increased transportion of weapons could cause increased probability of an explosive accident. It therefore is prudent to insure that our models provide adequate predictions of hazards to humans and their environment to study such potential accidents, and, in the event of an actual accident, to respond in real time.

One of the technical areas under investigation at the Lawrence Livermore National Laboratory in the USA, jointly with with the Institute of Experimental Meteorology and the Institute of System Studies in Russia, is the development of an automated methodology that insures effective utilization of the radiological measurements in conjunction with atmospheric

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dispersion modeling for real-time source term and dose estimation in the event of accidental release of radioactivity to the atmosphere. This work reports on the determination of MEDIC/MATHEW/ADPIC (1,2,3,4) model input parameters applicable to dispersion of ^{239}Pu due to a non-nuclear detonation of high explosive (HE).

If the HE charge in a nuclear weapon detonates and does not produce nuclear yield, much of the plutonium in the device is expected to be dispersed in the form of very fine particulate matter. To model such events, Foster, Freis and Nasstrom (5) have implemented in ADPIC an explosive "cloud rise" model developed at the Sandia National Laboratories by Boughton and Delaurentis (6). In particular, this model provides ADPIC with a time evolution of the physical and thermodynamic properties of a buoyant cloud. These include the cloud radius, R(t), height, z(t), temperature, T(t), and vertical velocity, W(t), as functions of time, t.

The ADPIC Lagrangian "mass" particles are randomly placed in the cloud at t=0. Subsequently, the particles are radially dispersed due to the expansion of the cloud, advect horizontally with the cloud, and receive a vertical component of velocity, $w(m_p,t)$, given by

$$w(m_p,t) = W(t)\exp\{-c[R_p(t)/R(t)]^2\} - s(m_p,z_p) \qquad 1)$$

where: m_p is the particle mass; $s(m_p,z_p)$ is the gravitational settling velocity of the particle at height z_p; R_p is the radial distance of the particle from the cloud center; and c is the particle "velocity coupling coefficient" which, for a positive value, causes those particles near the cloud center to receive more vertical lift due to the cloud rise that the lift received near the cloud edges. This model has been compared with a "stabilized cloud" model by Baskett and Cederwall (7). They concluded from sensitivity tests, that if an accurate dispersion calculation by the ADPIC model is required for Clean Slate 1, the time-dependent cloud rise model should be used.

In previous investigations (8,9) we developed a rudimentary automated methodology for estimating source release parameters that are optimal in the least squares sense of non-linear regression analyses. This approach represented a marked departure from the manual and time consuming method often used for source term and dose estimation. The methodology was designed to gain the maximum information on both the meteorological model (MEDIC and MATHEW) as well as the atmospheric transport and diffusion (ADPIC) model parameters. A prime objective was to develop the regression in a manner that is computationally efficient in order to do real-time predictions.

This paper reports on our implementation of the regression methodology using the "ARAC emergency response" version of ADPIC[a]. The viability of the approach is evaluated by estimation of ADPIC model input parameters of activity mean aerodynamic diameter, and geometric standard deviation[b]. Additionally we estimate an optimal velocity coupling coefficient. The experimental deposition data, normalized to 1 kg of ^{239}Pu, are taken from the Roller Coaster field experiments (10) conducted during 1963 at the Tonopah Test Range in Nevada.

2. PARAMETER ESTIMATION METHODOLOGY

For a given experiment that we wish to model, and for $i = 1,2,\ldots,I$, let $o_i \equiv o(\bar{x}_i)$ be a set of measured values (for example, the measured pollutant concentration) with uncertainties given by $\sigma_i \equiv \sigma(\bar{x}_i)$, at the independent variable $\bar{x}_i$ (where the independent variable may, in fact be a vector that represents, for example, a spatial location and a particular time for the measurement). Suppose the model fitting is dependent on varying a set of input parameters which are given by $\bar{\theta} \equiv \{\theta_j, \text{for } j = 1,2,\ldots,J\}$. Then, let $c_i \equiv c(\bar{x}_i,\bar{\theta})$ be the model computed concentrations in the same units as the measurements, and at the same independent (spatial and temporal) variables, $\bar{x}_i$.

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[a] The work reported in reference 8 used a "Sequential Gaussian Puff" model and that reported in reference 9 used a specialized version of ADPIC for sampling purposes.

[b] In addition to the truncated unimodal log-normal size distribution, we consider a bimodal distribution that is the equally weighted sum of two such distributions.

Then, for a particular choice of measurements, model, and model input parameters, we define a "least squares" goodness of fit, $\Phi = \Phi(\bar{x}_i, i = 1,2,\ldots,I; \theta_j, j = 1,2,\ldots,J)$, by

$$\Phi = \sum_{i=1}^{I}\left[w_i(o_i - c_i)^2\right] \tag{2}$$

where w_i is a weighting factor. Often used values for the weighting are $w_i = 1/\sigma_i^2$, or $w_i = 1/|o_i|$. Since our data may vary over orders of magnitude, and for some choices of parameters the computed concentrations may miss the data by several orders of magnitude, we have chosen to use the weighting factor defined by

$$w_i = R_i / |o_i| \text{ where } R_i \equiv \min\left\{10^5 o_i, \max\left[\frac{o_i}{c_i}, \frac{c_i}{o_i}\right]\right\}. \tag{3}$$

According to the method of least squares (c.f., 11,12,13), the optimum values of the model parameters $\bar{\theta}$ are obtained by minimizing Φ with respect to each of the parameters simultaneously. That is, we seek the set of parameters for which

$$\frac{\partial\Phi}{\partial\theta_j} = \frac{\partial}{\partial\theta_j}\sum_{i=1}^{I}\left[w_i(o_i - c_i)^2\right] = -2\sum_{i=1}^{I}\left[w_i(o_i - c_i)\frac{\partial c_i}{\partial\theta_j}\right] = 0 \tag{4}$$

for all of the θ_j. It general, it is not possible to derive an analytical expression for calculating all the partial derivatives for complex numerical models such as M/A. Instead, Φ must be considered a continuous function of the J parameters θ_j, describing a hypersurface in (J+1)-dimensional space which must be searched for the minimum value of Φ.

One of the difficulties of such a search is that for a non-linear function there may be more than one local minimum for Φ within a reasonable range of values for the parameters. Unless the range can be physically restricted to a region in which it is known that there is only one minimum (which we assume here), any of the so-called "gradient" methods may converge to a local rather than the global minimum.

Based on our earlier work (8) we have selected to implement the Marquardt (14) methodology which combines both a "steepest descent" i.e., Eq. (4), when far from the minimum, and a "first order Taylor's expansion of the fitting model parameters" as the minimum is approached.

3. DATA BASE: THE CLEAN SLATE 1 EXPERIMENT(10)

The Clean Slate 1 experiment was one of the Roller Coaster field experiments conducted during 1963 at the Tonopah Test Range in Nevada. The purpose of these experiments was to acquire the data needed to develop the predictive capabilities required to assess the environmental consequences of a nuclear weapon accident involving an explosive release of plutonium to the atmosphere due to an inadvertent detonation of the weapon's HE component without a nuclear yield. The experiments provided detailed information on the spatial and temporal distribution of the plutonium as a function of particle size in the cloud produced by the HE detonation as well as the distribution of the plutonium on the surface. The designs included (1) an extensive surface-based air and deposition sampling network situated along a series of arcs that extended in a cone out to a distance of about 10 km from the event site, and (2) a balloon-borne air sampling system that contained a large number of air samplers to define the vertical distribution of plutonium in the cloud. In addition, a comprehensive meteorological measurements program supported the experiments to define the surface and upper air wind flows and turbulence during the event.

Clean Slate 1 HE was single-pont detonated over a relatively flat area of the NTS test range to simulate an accident situation. The cloud rose quickly to a height in excess of 500 m due to its thermal buoyancy, and thereafter the fate of the plutonium was governed by gravitational settling, atmospheric transport and diffusion, and deposition on the ground surface.

Particle size measurements of the plutonium bearing particles indicated a significant variability of the size distribution as a function of height with the larger particles being associated with the lower part of the cloud. The sizes have been converted to equivalent diameters of unit density spheres. Estimates (10) using less formal methods have found the median diameter to be about 40 micrometers and that approximately 20% of the particles are respirable with diameters of less than 10 micrometers.

The upper level winds at the time of the event were fairly steady at 5 to 8 m/s with very little directional shear. The atmosphere was characterized as being relatively stable.

4. COMPUTATIONAL RESULTS

The MEDIC/MATHEW/ADPIC grids and winds have been chosen to agree with previously published results [7,15,16]. Based on Figure 3 of Baskett and Cederwall [7] which compares the observed to computed cloud top height, we initialize the explosive cloud at 50m above the surface. Case 1 initial values of activity size and coupling parameters, chosen consistent with the referenced computations, are input to begin the calculations. Initial values for Case 3 are just guessed based on the results of Case 2. The regression methodology then iterates to find the set of parameters where the least squares measure, Φ, converges to a local (and hopefully, a global) minimum.

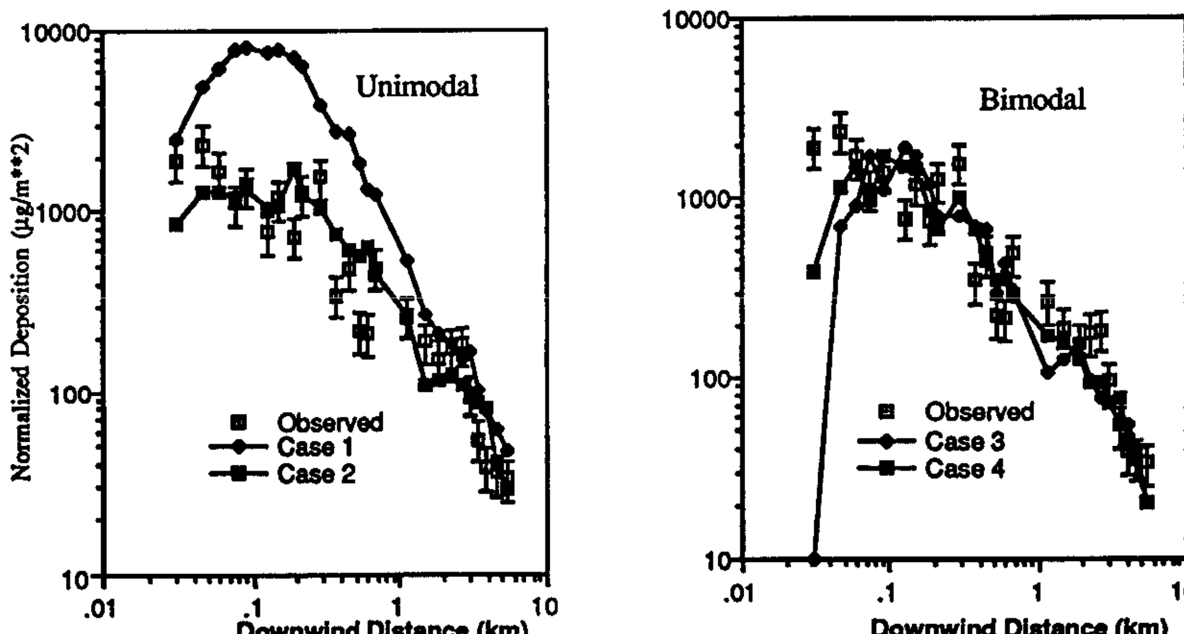

Figure 1: Observed and Model Computed Deposition (Normalized to 1 kg of ^{239}Pu) as a function of downwind distance from the source.

Figure 1 compares the deposition values as observed (with 25% error bars as estimated in (10)) and as model computed at differing distances downwind from the source. One observes that with Case 1, the model computed values are basically too high indicating that the activity size is too large. Case 2 is the model computations after a few regression iterations for the new particle size parameters and velocity coupling coefficient as shown in Table 1. Generally the fit to observed is significantly better as reflected in the table. Cases 3 and 4 compare the fits using a bimodal lognormal particle size distribution. The maximum and average factors of Case 3 are greatly inflated due to the fact that with the initial values of the parameters, the first sampler computes no deposition. Case 4 is after regression for the best fit parameters.

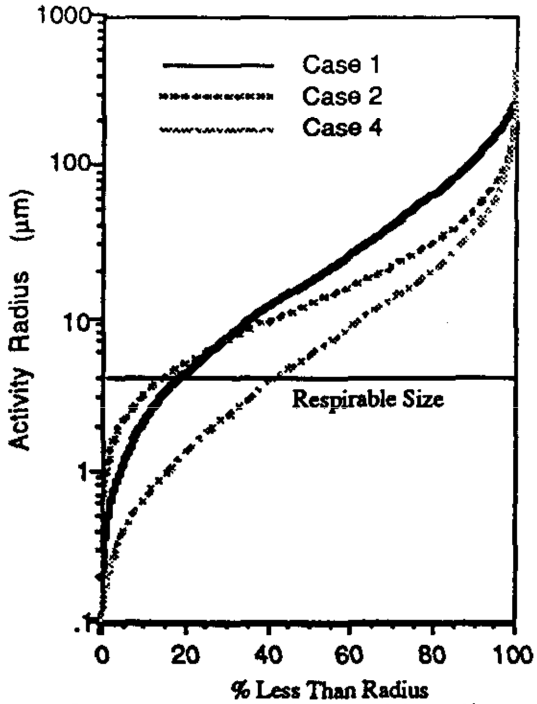

Figure 2: Percent of activity with radius less than the stated radius

Table 1 indicates the best overall fit by Case 4 based on the least squares value, the maximum factor and average factor of observed to computed. From Figure 1, even though the nearest to the source comparison with Case 2 may be worse, the Case 4 fit is clearly better beyond about 5 km.

Figure 2 displays the percent of marker particles with radius less than the radius given on the abscissa. We note that even though the median diameter is reduced by about 20% from Case 1 to Case 2, the fraction of respirable size particles is actually reduced only by a small amount due to the narrower distribution predicted by Case 2. The lowermost curve, Case 4, indicates almost a factor of 2 greater respirable fraction with the bimodal distribution. This distribution indicates a much greater hazard than previously reported for one-point HE detonations. This is, however, a preliminary result that warrants further investigation.

Table 1: Cases 1 and 3 are computed with "initial guesses" of the particle size parameters and coupling coefficient. Cases 2 and 4 show the improvements after a few regression iterations. Cases 1 and 2 use a truncated lognormal activity size. Cases 3 and 4 use a truncated, bimodal lognormal activity size. The maximum and average factors are the ratio of predictions to observations or vice-versa, whichever is larger.

Case	Median Radius (µm)	Maximum Radius (µm)	Geometric Std. Dev	Velocity Coupling	Maximum Factor	Average Factor	Φ
1	20.00	250	5.71	0.35	10.05	4.03	100.00
2	12.14	278	2.96	0.19	2.94	1.48	0.51
3	2.00, 15.00	350	2.00, 4.00	0.20	>200.00	1.57	>100.00
4	1.84, 14.01	437	3.70, 2.93	0.22	4.89	1.38	0.26

5. DISCUSSION AND CONCLUSIONS

From the figures and the table, one observes that, in general, the model suggests a smaller median particle size with a more narrow distribution than reported in [5,7,10,15,16], i.e., Case 1 with maximum radius of 100 µm. However, if the maximum is restricted to 100 µm, then no deposition is computed for the first 1/2 km downwind.

The purpose of implementing a good regression scheme is to best estimate the uncertain model input parameters so that the model can be used to make more accurate predictions outside the range of existing data or for subsequent model runs. These predictions, with estimates of their uncertainties, can then be used to determine actions recommended for emergency response. In our computations one observes a greatly improved fit to the observed deposition concentrations using the regression scheme. We note that with the "best fit" to deposition, the model also predicts the 3 highest observed air concentrations at the balloon curtain to within a factor of 2. This is considered an excellent fit by atmospheric dispersion modelers. However, the success of this technique is based on the premise that

a) the dispersion model is able to perform a credible simulation of the appropriate physical processes that are needed to account for the radiological observations; and,
b) the radiological measurements are sensitive to the effects of meteorology, topography and source characteristics on plume behavior.

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